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During the grant period, this work has focused on two research areas: zero electron kinetic energy (ZEKE) spectroscopy experiments on mass-selected cluster anions in which a halide ion (C1 or I) is solvated with a known number of rare gas atoms, and stimulated Raman pumping of negative ions. Negative ion ZEKE spectroscopy is a high resolution laser photodetachment experiment which yields information on the vibrational and electronic spectroscopy of both the anion cluster and the neutral species formed by photodetachment. By performing this experiment on a series of clusters (ArnI, for example), one learns how the stepwise solvation of the halide ion affects the anion and neutral energetics and spectroscopy. We have measured ZEKE spectra of ArnCl, ArnBr, ArnI, and XenI clusters. In all cases, we obtain high quality intermolecular potentials for the diatomic neutral and anionic (i.e. n=1) clusters, and learn about the how the spin-orbit levels of the neutral halogen atom are perturbed by the rare gas atom. We have also performed stimulated Raman pumping in C2, the first such observation for a molecular negative ion.

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Daniel M. Neumark AFOSR/AASERT Grant Number F49620-94-1-0412 Photo-Initiated Ion-Molecular Reactions in Size-Selected Clusters

FINAL TECHNICAL REPORT

AASERT funds have been used to support the research of a graduate student, Mr. Ivan Yourshaw, in conjunction with his efforts on my primary AFOSR grant (FR49620-94-1-0115). During the grant period, this work has focused on two research areas: zero electron kinetic energy (ZEKE) spectroscopy experiments on mass-selected cluster anions in which a halide ion ($C\Gamma$ or Γ) is solvated with a known number of rare gas atoms, and stimulated Raman pumping of negative ions. Negative ion ZEKE spectroscopy is a high resolution laser photodetachment experiment which yields information on the vibrational and electronic spectroscopy of both the anion cluster and the neutral species formed by photodetachment. By performing this experiment on a series of clusters ($Ar_n\Gamma$, for example), one learns how the stepwise solvation of the halide ion affects the anion and neutral energetics and spectroscopy.

Thus far we have measured ZEKE spectra of Ar_nCl̄, Ar_nBr̄, Ar_nΓ, and Xe_nl̄ clusters. In all cases, we obtain high quality intermolecular potentials for the diatomic neutral and anionic (i.e. n=1) clusters, and learn about the how the spin-orbit levels of the neutral halogen atom are perturbed by the rare gas atom. Using this information, we can then deduce the nature and extent of non-additive forces that play a role in the n>1 clusters. We find the largest effects to be in the anion clusters, primarily through induction (i.e. the interaction between neighboring charge-induced dipoles) and exchange-induced quadrupole effects in the solvent atoms. Previously, many-body effects in clusters have been studied through the observation of small spectral shifts in high resolution spectra. Our results directly yield the solvent binding energies in these clusters, and this appears to be a clearer probe of non-additive forces in clusters.

The Ar_nCl^- experiments are of particular interest because our results can be directly compared to detailed theoretical treatments of these clusters. The $Xe_n\Gamma$ experiments are of interest in light of recent experiments by Cheshnovsky and co-workers in which the cluster analog of the charge-transfer-to-solvent (CTTS) band has been observed for these species. Our experiments will indicate whether this band lies above or below the adiabatic detachment energy for these clusters, a vital piece of information in the interpretation of the CTTS studies.

We have also performed stimulated Raman pumping in C_2 , the first such observation for a molecular negative ion. In stimulated Raman pumping, a vibrational transition is pumped by two lasers, one tunable and one fixed in frequency, whose frequency difference equals the energy of the vibrational transition. In this case we pump the v=0, N=2 level of C_2 to the v=1, N=2 level, and use resonant two-photon detachment (R2PD) to probe either depletion of the v=0 level or gain in the v=1 level resulting from stimulated Raman excitation. This bodes well for the primary goal of this research, in which stimulated Raman pumping will be used as a general means of vibrationally exciting negative ions and measuring their photoelectron spectra.